

Kinetic Monte Carlo simulations of self organized nanostructures on Ta Surface Fabricated by Low Energy Ion Sputtering

Shalik Ram Joshi¹, Trilochan Bagarti², and Shikha Varma^{*1}

¹Institute of Physics, Sachivalaya Marg, Bhubaneswar-751005

²Harish-Chandra Research Institute, Chhatnag Road, Jhansi, Allahabad-211019

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Abstract

Surfaces bombarded with low energy ion beams often display development of self assembled patterns and quasi-periodic structures. Kinetic Monte Carlo simulations have been performed to describe ion sputtered Tantalum surfaces. A weak nonlinearity in the relaxation process has been introduced and the results show that the Positive Schwoebel barrier, produced by the nonlinear Hamiltonian, is necessary in describing ion bombarded Tantalum surfaces. Furthermore, their scaling exponents suggest presence of a class other than KPZ.

1 Introduction

Non-equilibrium surfaces produced via bombardment with energetic ion beams, often exhibit well ordered patterns having several potential applications [1–3]. The surface morphology, here, develops as a consequence of competition between a variety of processes like roughening dynamics, relaxation processes, generation of defects, material transport etc. [4, 5]. Occurrence of a wide array of surface morphologies complicates the prediction of dominant mechanism controlling their evolution. Non-metallic surfaces generally display hills and depressions for normal incident ion beams [6], while showing quasi-periodic ripple morphology under off-normal conditions [7, 8]. For metallic surfaces, ripples develop even at normal incidence [9]. Symmetry breaking anisotropy in surface diffusion can cause these ripples to rotate with substrate temperature [9, 10]. For low energy ion beam induced patterning, although the erosive processes are dominant, enhanced surface diffusion due to the defect mobility also becomes important especially under the low ion-flux conditions [4, 7]. The surface morphology in this scenario is governed by the non-equilibrium biased diffusion current.

*shikha@iopb.res.in

Continuum model approach has proved to be a very successful technique in describing the surface evolution but involvement of several complex phenomena make it difficult to relate with the experimental results [11]. An alternative approach is the Kinetic Monte Carlo(KMC) method where the kinetic behavior of the surface is simulated at microscopic level for a discrete surface. During diffusion or sputtering, the surface gets modified in the units of whole atoms, only at the specific site [12]. Various models have been proposed to understand the effect of sputtering related erosion on surface evolution. For instance, in model by Cuerno et.al [13], the local surface morphology has been used to determine the sputtering yield of the surface, while in binary collision approximation, the atom gets removed from the surface with a probability proportional to the energy deposited, in its near surface region, by incoming ions [14–16]. Surface diffusion has been formulated via relaxation of the surface to its minimum energy through a series of atomic jumps with probability that depends upon the energy of the initial and the final state [17]. Thermally activated hopping, where an atom hops over an energetic barrier with a barrier height that depends on the local configuration, has also been considered for studying surface diffusion [14, 16, 18].

In the present article, KMC results are presented in 1+1 dimension for ion beam modified metallic surfaces. Experimentally, the presence of ripple morphology on ion irradiated Tantalum surfaces has been observed [19,20]. We have developed a model based on earlier work by Cuerno *et. al.* [13] which was not able to describe the surface morphology of ion sputtered metallic Tantalum surface. The Schwoebel effect is found to be important for Tantalum surface, and has been incorporated in our model, by including a weak non-linearity in Hamiltonian for relaxation of diffusing atoms on the sputtered surface. Simulation results, presented here, shows that the presence of Schwoebel effect produces the surface morphology and scaling exponents that are consistent with our experimental observations. The scaling exponents indicates that the morphology of ion sputtered Tantalum surfaces may belong to universality class other than Edward-Wilkinson(EW) and Kardar-Parisi-Zhang(KPZ). The paper is organized as follows. In Section 2 we describe the KMC model. The experimental details are described in Section 3 and in Section 4 we discuss the results.

2 The Model

In our model we assume that ion irradiation causes the surface to evolve by two dominant processes, namely erosion and surface diffusion. The erosion process consists of the removal of atoms from the surface due to the impinging ions. Ions bombarded on the surface penetrate deep into the substrate releasing energy to the neighboring atoms along the trajectory. If the energy gained by a surface atom is sufficiently large, it gets eroded from the surface. Sputtering yield, $Y(\phi)$, defined as the number of atoms eroded for every incoming ion at an angle ϕ to the surface normal. It depends on the energy of the ion and local morphology of the interface and, is a measure of the efficiency of the sputtering process [21]. We assume $Y(\phi) = a + b\phi^2 + c\phi^4$, where a, b and c are constant such that $Y(\pi/2) = 0$ and for some critical ϕ_0 , $Y(\phi)$ has a maxima. The

erosion process brings in an effective negative surface tension that causes the surface to become rough [22].

Surface diffusion, on the other hand, consists of the random migration of surface atoms on the surface such that the surface energy is minimized. Its strength depends on the temperature of the substrate and the binding energy of the atoms. The negative surface tension, during erosion, leads the system towards instability and as a result the system responds to restore stability by surface diffusion [13].

We consider a one dimensional lattice with periodic boundary conditions. The surface at any instant of time t is described by the height $h_i(t)$ at each lattice site $i = 1, \dots, L$. We consider initially (at time $t = 0$) a flat surface $h_i(0) = \text{const.}$ The erosion takes place with probability p while the diffusive process occurs with probability $(1 - p)$ at a randomly chosen lattice site. The surface is evolved by following dynamical rules.

(i) *Erosion*: The particle on the surface at site i gets eroded with a probability $Y(\phi_i)p_e$ where $\phi_i = \tan^{-1}((h_{i+1} - h_{i-1})/2)$ and $1/7 \leq p_e \leq 1$ is the ratio of the number of occupied neighbors to the total number of neighboring sites [21]. The ratio p_e accounts for the unstable erosion mechanism due to the finite penetration depth of the bombarding ions into the eroded substrate [13, 22, 23].

(ii) *Surface diffusion*: The surface diffusion process is taken into account by nearest neighbor hopping. The hopping rate from an initial state i to a final state f is given by $w_{i,f} = [1 + \exp(\beta\Delta H_{i \rightarrow f})]^{-1}$ where $\Delta H_{i \rightarrow f} = H_f - H_i$ is the difference in the energy of the states and $\beta^{-1} = K_B T$ where T is the surface temperature and K_B is the Boltzmann constant. The surface Hamiltonian is given by

$$H = \frac{J}{2} \sum_{\langle i,j \rangle} |h_i - h_j|^2, \quad (1)$$

where $\langle i, j \rangle$ denotes sum over nearest neighbor sites and J is the coupling strength. Similar relaxation behaviour has been considered by Cuerno *et. al.* [13] to describe the surfaces evolving from initial ripple structure into rough surfaces of KPZ class. Morphology of several other non-metallic surfaces, post ion irradiation, have also been describe sufficiently well by this model [24, 25].

The morphology of metallic surfaces, however, cannot be described by the above model alone. In these cases, a diffusing atom is repelled from the lower step and preferably diffuses in the uphill direction. This Schwoebel effect is controlled by the potential barrier, Schwoebel barrier, and has been considered to be crucial for MBE grown surfaces [17, 26, 27]. Growth models based on MBE studies, have shown that positive Schwoebel effect can be incorporated in the relaxation process Hamiltonian by the inclusion of a quartic term [17]. In the present study, we model the relaxation behaviour on ion sputtered Tantalum metal surfaces by modifying Eq. (1) to include the Schwoebel effect in the Hamiltonian:

$$H = \frac{J}{2} \sum_{\langle i,j \rangle} |h_i - h_j|^2 + \epsilon |h_i - h_j|^4. \quad (2)$$

Here $0 < \epsilon < 1$ is a non-linearity parameter which controls the intensity of Schwoebel effect. The additional quartic term in Eq. (2), as the results presented here shows, can be crucial for relaxation after ion irradiation of metallic surfaces as it is responsible for an uphill current which results in the formation of sharp peaked 'groove' structures.

The algorithm for the Monte Carlo simulation is following. A site $1 \leq i \leq N$ is chosen at random and is subjected to follow erosion process with probability p or the diffusive process with probability $1 - p$. If erosion process is chosen, the angle ϕ_i is computed and a particle is eroded with probability $Y(\phi_i)p_e$. On the other hand if diffusive process is chosen, $w_{i,f}$ is computed using the surface Hamiltonian Eq. (2) and the new configuration is updated. Time t is incremented by one unit.

3 Experimental Details

High purity (99.99%) Tantalum foils were bombarded by 3keV Ar ions under UHV conditions. The angle of incidence for ion beam was 15° w.r.t surface normal and its flux was 10^{13} ions/cm². Scanning Probe Microscopy (SPM) studies have been conducted on the surfaces by using Bruker (Nanoscope V) system in tapping mode.

4 Results and discussion

Fig. 1 displays an SPM image from a Tantalum surface bombarded by Ar⁺ ions at fluence of 3.6×10^{16} ions/cm². A quasi periodic ripple pattern with a wavelength of ~ 80 nm is observed. 1-dimension height profile from the experiment (section marked in Fig. 1) and from KMC simulations are presented in Fig. 2.

For KMC simulations, first we study the model that considers erosion and includes relaxation mechanisms via only quadratic term in the Hamiltonian i.e for $\epsilon = 0.0$ in Eq. (2). A range of parameters were chosen for simulations and the results are presented in Fig. 2, for $p = 0.1$, $J_c/K_B T = 0.25$. The height profile shows a periodic structure, usually similar to the morphologies observed for non metallic surfaces [28] where the Schwoebel effects are not essential. Experimental height profile has several sharp peaks and grooves while the simulated profile has only smooth morphology.

Next we examine the model where we consider relaxation of the surface by including both quadratic and quartic terms in the Hamiltonian Eq. (2). The non-linear parameter ϵ is varied between 0.001 and 1.0. The surface morphology with $\epsilon = 0.01$ is presented in Fig. 2 for $p = 0.1$, $J_c/K_B T = 0.25$. The simulated height profile here, with $\epsilon = 0.01$, displays good agreement with experiments where formation of groove like structures are clearly observed. These features are characteristic signature of positive Schwoebel barrier that force the atom to move in uphill direction by breaking the translational invariance symmetry. A high diffusion rate, as observed here ($p = 0.1$), is expected for metallic surfaces [10]. The steady state height profile for KMC simulations are also shown in Fig. 2 (inset).

The scaling behaviours and related exponents have also been explored here to investigate the nature of the growing surface. The exponents are useful as they depend on the growth condition and not on the microscopic details of the system. The correlation length ξ , which represents the typical wavelength of fluctuations on the growing surface, also characterizes the phenomenon of kinetic roughening. The width of the surface grows as $W(t) \sim \xi(t)^\alpha$ for roughness exponent α . The scale invariant surfaces lead to scaling laws for correlation functions. The equal time height-height correlation (HHC) function can be written as:

$$G(\mathbf{r}, t) = L^{-1} \sum_{\mathbf{r}'} \langle [h(\mathbf{r} + \mathbf{r}', t) - h(\mathbf{r}', t)]^2 \rangle. \quad (3)$$

Here \mathbf{r} is the Translational length along lateral direction of the 1-d lattice and $\langle \cdot \rangle$ denotes the ensemble average. This HHC function has the following scaling form:

$$G(\mathbf{r}, t) = r^{2\alpha} g(r/\xi(t)). \quad (4)$$

with $g(x) \sim \text{constant}$ for $x \ll 1$ and $g(x) \sim x^{-2\alpha}$ for $x \gg 1$

Fig. 3 presents the 1-dimensional height-height correlation function for experiment (using Fig. 1) as well as KMC simulations. By utilizing the phenomenological scaling function of form $H(r) \sim [1 - \exp(-(r/\xi)^{2\alpha})]$, values of ξ and α have been obtained and are listed in Table 1. Although in absence of any Schwoebel effect ($\epsilon = 0.0$), the simulation results are very different from experimental HHC function, after inclusion of Schwoebel effect ($\epsilon = 0.01$) the results are in agreement. These results demonstrate that a small nonlinearity parameter with $\epsilon = 0.01$ is essential for achieving experimentally consistent HHC functional form, ξ and α . This indicates that Schwoebel effect is necessary for understanding correct growth behaviour on Tantalum surface. Value of α obtained here for KMC simulation, in absence of Schwoebel effect ($\epsilon = 0.0$), is similar to that observed in literature for MBE models with linear Hamiltonian [29, 30].

	α	ξ
Experiment	1.22 ± 0.26	5.66 ± 0.36
$\epsilon = 0.0$	1.63 ± 0.35	2.63 ± 0.50
$\epsilon = 0.01$	1.20 ± 0.07	7.14 ± 0.05

Table 1: Roughness exponent α and correlation length ξ .

Obtaining α from $G(\mathbf{r}, t)$ can be difficult when the correlation length reaches the system size, specially in the steady state regime. In order to neglect finite size effects α can be computed in $0 \leq r \leq L/2$ regime, where L is the system size. Structure Factor mentioned below does not encounter this problem. The Structure Factor can be defined as [17]:

$$S(\mathbf{k}, t) = \langle \hat{h}(\mathbf{k}, t) \hat{h}(-\mathbf{k}, t) \rangle. \quad (5)$$

Here, $\hat{h}(\mathbf{r}, t) = L^{-d/2} \sum_{\mathbf{r}} [h(\mathbf{r}, t) - \bar{h}] e^{i\mathbf{k}\mathbf{r}}$, is the associated correlation function and \bar{h} is the spatial average of $h(\mathbf{r}, t)$. This function has the following scaling form:

$$S(\mathbf{k}, t) = k^{-\gamma} s(\mathbf{k}^{zt}). \quad (6)$$

with $\gamma = 2\alpha + d$. The scaling function s approaches a constant for large argument but behaves differently in the short time limit $x \ll 1$ where it has form

$$s(x) \sim \begin{cases} x & \text{if } \gamma \leq z \\ x^{\gamma/z} & \text{if } \gamma \geq z \end{cases} \quad (7)$$

In Fig. 4, the steady state structure factor $S(\mathbf{k}) = S(\mathbf{k}, t \rightarrow \infty)$ is shown for experiment as well as from KMC simulations. The result clearly demonstrate that the non-linear Hamiltonian, with $\epsilon = 0.01$, agrees quite well with the experimental results. For $\epsilon = 0.0$, we observe that $S(\mathbf{k})$ qualitatively differs from the experimental results. The value of exponent γ obtained by linear Hamiltonian is 4.00 ± 0.12 while for non-linear Hamiltonian, the value is 3.27 ± 0.07 . For the ion beam modified Tantalum surfaces, $\gamma = 3.04 \pm 0.06$ has been observed here. This value of γ is not expected from the universality classes of EW or KPZ. Thus the results indicate that positive Schwoebel effect is necessary for understanding the morphological growth of ion sputtered Tantalum surfaces which does not belong to the well known EW or the KPZ class.

5 Conclusion

In Conclusion, we have presented a KMC model to describe the morphology of ion sputtered Tantalum surfaces. We have shown that a positive Schwoebel effect is needed to explain the characteristics of self organized nanostructures observed in the experiment. The Schwoebel effect has been used earlier for MBE growth models. Here, we have shown that, it can also be used for ion sputtered metallic surfaces such as Tantalum. The scaling exponents computed from the simulation and experimental data agrees quite well and indicates the presence of universality class that differs from that of non-metallic surfaces.

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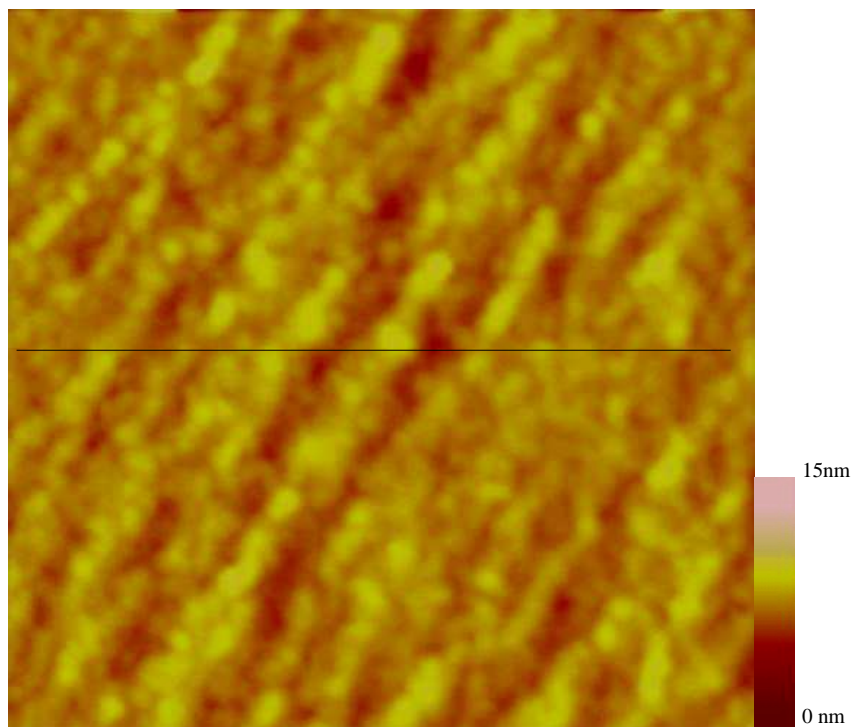


Figure 1: Morphology of Tantalum surface: SPM image ($500\text{nm} \times 500\text{nm}$) after ion beam irradiation at a fluence of $36 \times 10^{15} \text{ions/cm}^2$.

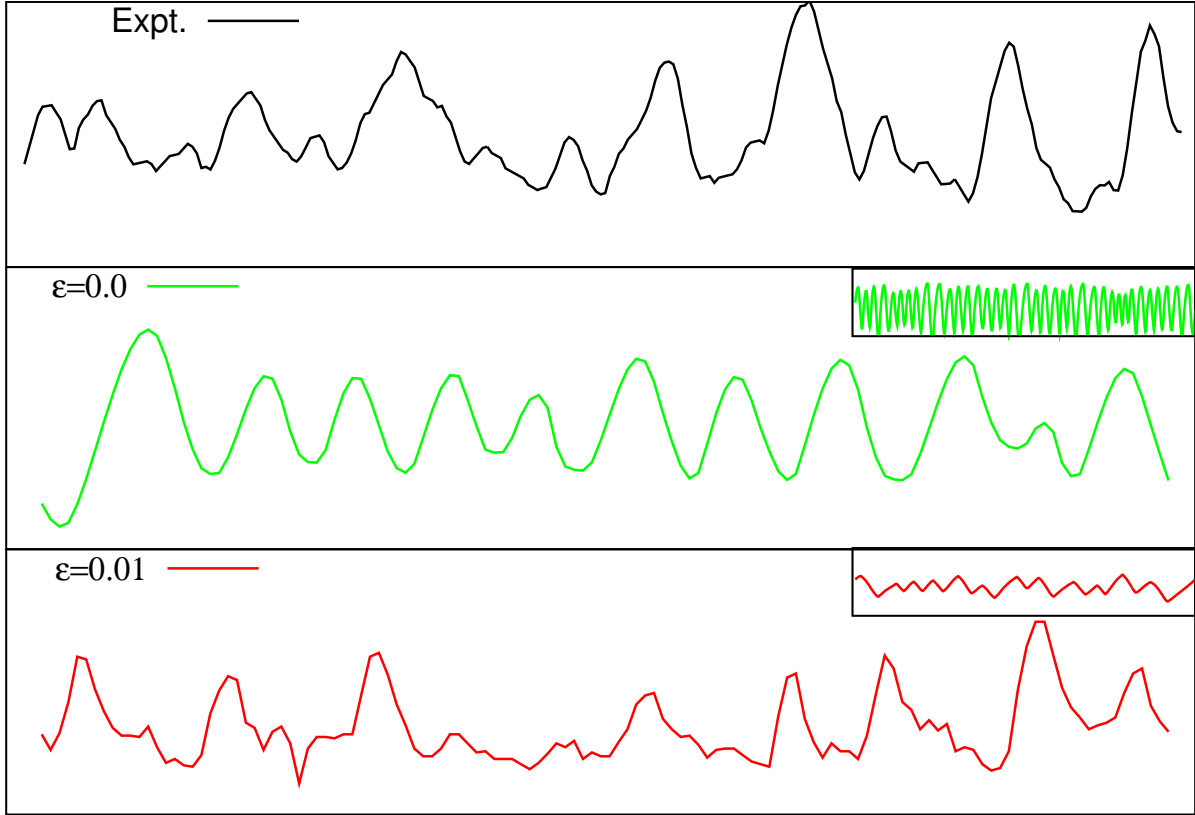


Figure 2: Comparison of surface profile($h(x,t)$) of experiment with the simulations. The simulated profiles were obtained by KMC model with the parameters $p = 0.1$, $J_c/k_B T = 0.25$ and $\epsilon=0.0$ or 0.01 . Inset shows the steady state profile of the surface

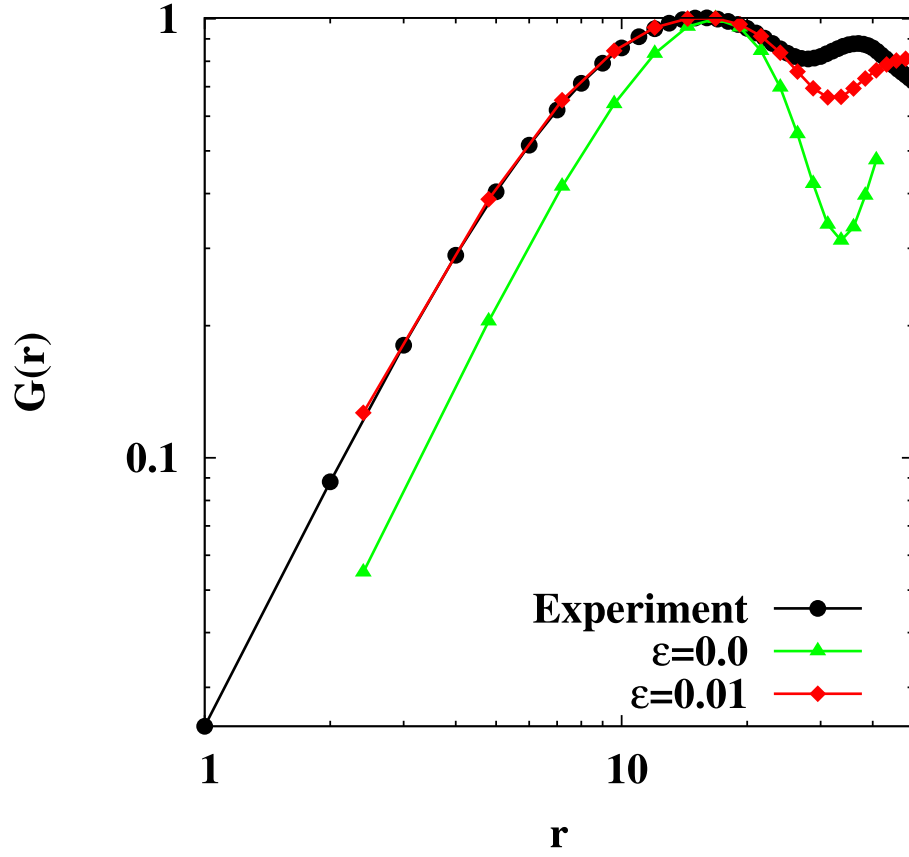


Figure 3: Equal time HHC $G(r,t)$ as a function of translational distance r for experiment. Simulation results are presented (for same parameters as in Fig. 2) with $\epsilon=0.0$ or 0.01 .

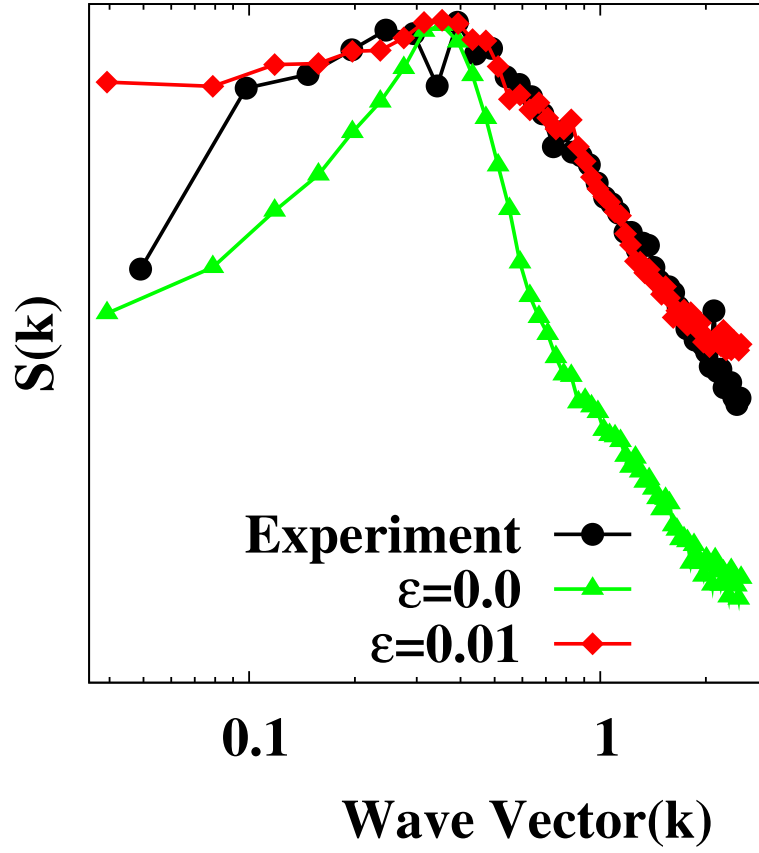


Figure 4: Steady state structure factor $S(k)$ for experiment. KMC simulation results are presented (for same parameters as in Fig. 2) with $\epsilon=0.0$ or 0.01 .